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Introduction to Separation Index: Modelling of Rare Earth Element Extraction Complexity for Feasible Processing

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Abstract

With the increasing demand from industries, the efficiency as well as the sustainability in extracting Rare Earth Elemnt (REE) from its main resources (mainly mining) has become very challenging. However, there is no specific study has been conducted particularly in optimizing the sequence of the extraction steps or reducing the complexity of separation stages when there are multiple choices of REE materials can be potentially extracted. In this light, this paper investigates the suitability of using the three main contributors of any Solvent Extraction (SX) system – separation factor value, material composition and number of processing stages (extraction and scrubbing), in specifically justifying the potential feasibility of the extraction pathways that has been initially selected for separation processing. In particular, the concept of Separation Index (SI) is introduced in correctly balancing the three corresponding factors of SX system, where 'Art vs Science' structure was used as the primary separation framework. In addition, two sets of Separation Factors - HDEHP-HCL and RE(III)-HCL-EHEHPA systems, are utilised to demonstrate the SI application. The results generally indicate that the proposed SI has sufficiently harmonizing the three main parameters of SX system, by which the low separation difficulty consists of the following criteria - relatively high SF value, relatively high material composition and relatively low number of stages. Through this work also, the 'Árt vs Science' is found favour the inter REE extraction rather than the intra separation. This is due to the chosen extractants generally shows low SF values mostly within the individual REE groups – light, medium and heavy REEs, respectively.

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Keywords: Rare earth separation, solvent extraction, separation factors, Art vs Science separation structure

1. INTRODUCTION AND MOTIVATION

Rare earth element (REE) is a unique group of metals which consists of 15 elements of lanthanides as well as scandium and yttrium. Within the lanthanides group, the elements can be further categorized into light (La, Ce, Pr, Nd), medium (Sm, Eu, Gd) and heavy (Tb, Dy, Ho, Er, Tm, Yb, Lu) groups (J. Zhang & Zhao, 2016). This classification is based on their solubility in double salt or extractability towards acidic extractant. Their main role is very crucial particularly in various manufacturing industries comprising of industrial magnet, catalyst, glass polishing, phosphors (electronic devices), ceramics as well as defence. All REEs are mainly recovered from mining processing which typically involves five major steps – mining (from the ground to crushed ore), milling (beneficiation), hydrometallurgical processing (transforming REE minerals into RE oxides), separation (extracting as well as purifying the individual REE), refinement (adjusting the REEs quality according to a specific technological demand specification) prior to downstream processing

(Golev, Scott, Erskine, Ali, & Ballantyne, 2014).

With the increasing demand from industries, the efficiency as well as the sustainability in extracting REE from its main resources (mainly mining) has become very challenging. This work particularly concentrates on the separation stage, where it is found that the 'solvent extraction' (SX) system has been identified as the most widely applied by the RE industry as commented in (Xie, Zhang, Dreisinger, & Doyle, 2014). However, there is no specific study has been conducted particularly in optimizing the sequence of the extraction steps or reducing the complexity of separation stages when there are multiple choices of REE materials can be potentially extracted. The current approach is solely adopting the separation factor as the sole basis in prioritizing the REEs extraction processing (Cheng et al., 2014; Morais & Ciminelli, 2004). Thus, the current industrial REEs are designed, in such a way perhaps, only a specific set of REEs of interest are solely to be recovered which corresponding to the pre-specified economical or demand constraints.

In this light, this paper attempts to explore the suitability of using the three main components of any SX extraction system – separation factor value, material composition and number of processing stages (extraction and scrubbing), in specifically justifying the potential feasibility of the extraction pathways that has been initially selected for separation processing. In other words, this work strives to formulate a specific parameter whereby the recovery process complexity is reduced by correctly balancing the high concentration of extracted materials with minimized number of stages. In order to achieve this, the following objectives are crucial to be addressed:

1. To introduce the concept of Separation Index (SI) particularly to determine the priority of rare earth element (REE) extraction sequence.
2. To demonstrate the SI application based on the processing structure of common REE processing sequence (Art vs Science).
3. To analyse the separation complexity of REE based on two sets of separation factor configurations.

2. RARE EARTH SOLVENT EXTRACTION PROCESSING

Solvent extraction is an important technique that is usually employed to separate and extract individual metals or to get their mixed solutions and compounds. The rare earth produced from different ores or solutions is a mixed product containing various individual rare earth elements. Basically, rare earth ore minerals will be through beneficiation process which is required for removing impurities and other radioactive element. After beneficiation process, the rare earth will be through leaching as well as separation process and finally to be market for high technology application. In the industry, rare earth producer starts by separating different groups of rare earths from the leachate (light, medium and heavy groups). Depending on the clients need, some primary rare earths producers may stop the process and sell the product as intermediate, mixed products, or perform different downstream separations to produce individual rare earth salts or oxides. However, to produce an individual rare earth oxide with high purity are difficult due to their similar physical and chemical properties. These process required up to hundreds separation stages and used high volume of extractants as well as acid.

Solvent extraction chose as the most practical method for rare earth as it offers simpler extraction setup with lower cost for a larger amount of ores (Peppard, Mason, Maier, & Driscoll, 1957; Peppard et al., 1957). However, before the booming of industrial scale solvent extraction in the 1960s, ion exchange technology was the only practical way to separate the rare earths in large quantities. Ion exchange however still be used nowadays to obtain small quantities of high purity rare earth especially for electronics or analytical applications. A basic solvent extraction process includes extraction, scrubbing/ washing, and stripping stages. Like the solvent extraction of other metals, the solvent extraction of rare earth exploits the separation of REEs between two immiscible liquids to perform the separation of one or one group of rare earth elements from the others (Figure 1). An aqueous phase usually acid loads with rare earth metals and mix with organic phase or "extractant" in extraction stage. The interest RE is denoted by red shaped object and others with green. Based on the abilities of the RE to distribute themselves between an aqueous solution and an immiscible organic solution, certain REE are selectively shift into the organic phase. The extracted RE are depend on the amount of H^+, with the highest amount separates the lanthanide series further to the

right on the Periodic Table (Kronholm, Anderson, & Taylor, 2013). The extraction process usually repeated few times to increase the separation of the interest rare earths towards other metals. At the end of the extraction process, the impurities that stays in the aqueous phase are channel to raffinate output and the extracted RE in the organic phase that consists of targeted RE and some impurities are sent to the scrubbing process. Following the extraction is scrubbing process. This process is carried out by adding water or dilute acid / base solution into the pregnant organic phase to remove any impurities or unwanted metals. This process is proven to improve the purity of the desired metals. The organic phase that contains only desired RE are stripped by dilute acid or base and the organic phase is recycled back to the extraction stage. The pregnant stripping solution leaves stripping point with a high concentration of RE and will be recovered as RE oxide after a series of downstream processes.

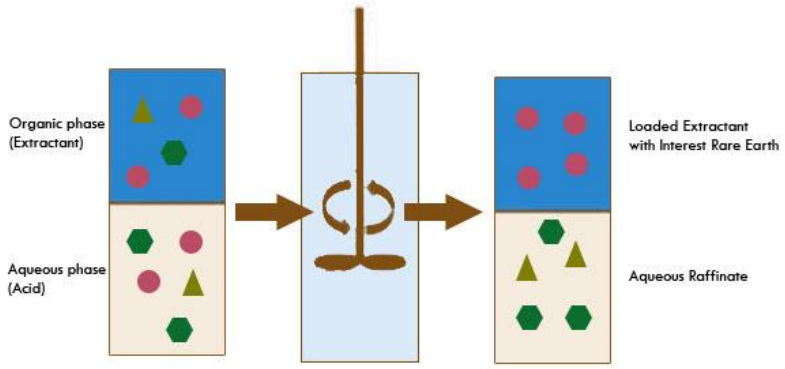


Figure 1: Simplified schematic of rare earth extraction process

3. THE CONCEPT OF SEPARATION INDEX

In the current literature, there is no specific study has been focusing on analysing the separation complexity particularly for REEs. However, it is generally understood that there are several factors simultaneously affect the efficiency of SX system including the type and concentration of extractant as well as acid used, the synergistic and coordination equilibrium effect. The following Table 1 presents a number of studies which discuss the impact of those factors on the REEs separation processing.

Table 1: Several contributory factors of SX system

No.	References	Extraction Factors/ Scopes	Findings
1	(Chang, Li, Liu, Hu, & Zhang, 2010; Wu, Zhang, & Bao, 2007; Xie et al., 2014)	Type of acid and extractant, equilibrium time, pH of the medium and concentration of the extractant	Most of the RE portrayed better extraction in the acidic medium. P204 and P507 considered as the most versatile extractant for RE extraction.
2	(Luo, Huang, Zhu, Long, & Liu, 2009; Xiaobo Sun, Meng, & Li, 2006; Xiaoqi Sun & Waters, 2014; C. Zhang et al., 2014)	Synergistic effect	Combination of P204/P507 with other extractant usually used to improve the extraction with enhancement coefficient of 1 – 5.
3	(Chen, Chen, Jing, & Li, 2015)	Coordination equilibrium effects	Better RE-extractant symmetry and stability significantly improve the extraction and selectivity of the process.

All these factors are identified not only influence the separation process as a whole but they are also directly associate to each other significantly. For instance, both separation factor values as well as the material composition are utilised in calculating the separation complexity as reported in (J. Zhang & Zhao, 2016). In this regard, this study employed both factors for representing the separation complexity in relation to REE processing. In particular, equation 1 shows the mathematical formula of separation index that proposed.

$$SI = \frac{F_{sf}}{N} \tag{1}$$

Where, F_{sf} = Scale factor (refer to section 5.0 later)

N = number of stages (refer to section 5.0 later)

From equation 1, the numerator signifies the scale factor which derived from the multiplication between the particular separation factor score and material composition corresponding to the specific individual or group of REEs that intended to be extracted (the details are provided in section 5). Meanwhile, the denominator relates to the number of stages which calculated from the concurrent SX operation modelling that developed in (J. Zhang & Zhao, 2016). By conception, equation 1 works almost similar to the function of separation factor. In particular, if the SI value is relatively high, it basically indicates that the separation difficulty is somewhat less. In other words, if the SI value is very close or less than one (1), by theory it shows that the particular REE extraction is rather complex. In this respect, thus, the concept of separation complexity or difficulty that symbolised by SI in this study is directly indicates one or the combination of these criteria.

i. Relatively low SF value. When the SF value is low, then the chosen extractant is facing huge difficulty in separating the specified REE from the crude material. The fundamental details of SF can be obtained in (J. Zhang & Zhao, 2016).
ii. Relatively low material composition. If the targeted REE composition is low, then it is not feasible to conduct the separation operation economically. If insists, it may lead to great financial loss even the demand is consistently increasing.
iii. Relatively high number of separation stages. In this particular situation, applying large operation stages may increase the cost of operations as other operation variables such as the amount of extractant, utilities, man power as well as instrumentation may also rise concurrently.

In every of those situations, the term 'relative' is applied because there is no specific value which can be used as a standard in differentiating between the difficult and feasible separation. Hence, the SI value is not a constant for every individual REE in all separation contexts. In summary, the SI magnitude is very much dependant on the scale of SF configuration, the proportion of that particular REE extracted and also the calculated total number of separation stages.

4. REE SEPARATION BASED ON 'ART VS SCIENCE' STRUCTURE

The 'Art vs Science' processing structure by (Mackowski, 2014) is chosen as the base case for demonstration as portrayed in Figure 4. From Figure 4, the process covers comprehensively all sorts of REEs separation which can be subdivided into three main pathways – light (LREO), medium (MREO) and heavy (HREO) groups respectively as listed in Table 4.

From Table 2, the first step specifically separates MREO and HREO as a bulk from the original raw material input. In the second step, the focus is on extracting the specific elements of MREO from all the HREO components. In particular, from Table 2, steps 2a and 2b concentrate on the individual extraction of MREO REEs starting with Gd and followed by Eu as well as Sm. Meanwhile, steps 2c and 2d correspond to the separation of HREO element. In this regard, however, the original description mainly highlights recovery of Y and Dy only, while leaving the other HREO components as residue. This is perhaps due to the proportion of HREO is typically small and it is not economically viable for recovery. The last separation stage (step 3) primarily focusing on LREO separation. In this particular

phase, the separation operation can be divided into two sub-series – recovery of Pr and Nd in one hand, while separating on Ce from La on the other. Therefore, pre-extracting of Pr and Nd as a group, which also called as 'Didymium' is initially conducted. This operation also is practically reasonable because Didymium is known for its unique magnetic character, and thus, the separation mechanism should be designed exclusively. In addition, both Ce and La are usually in high composition, hence, if the Didymium is not effectively removed at the beginning of step 3, then the complexity of attaining high purity of Ce or La at the later stage is also relatively great as well as higher separation stages are expected.

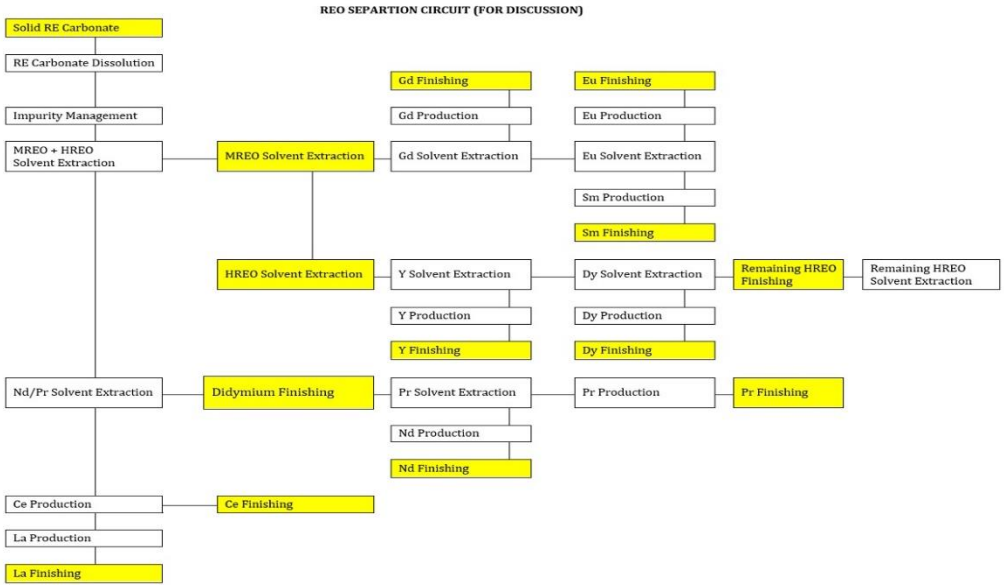


Figure 2: Separation pathways of REEs based on 'Art vs Science' structure (Mackowski, 2014)

Table 2: Individual REEs Extraction Step based on 'Art vs Science' Structure

Steps	Description	REE Involvement
1	Separation of MREO+HREO from the raw mat	Ce,La,Pr,Nd,**Sm,Eu,Gd,Dy,Ho-Lu**
2	Separation of MREO from HREO	**Sm,Eu,Gd**,Dy,Ho-Lu
a	Extraction of Gd from Sm,Eu	Sm,Eu,**Gd**
b	Extraction of Eu from Sm	Sm,**Eu**
c	Extraction of Y from Tb-Lu	**Y**,Tb-Lu
d	Extraction of Dy from Tb,Ho-Lu	**Dy**,Tb,Ho-Lu
3	Separation of Nd/Pr from Ce-La	La,Ce,**Pr,Nd**
a	Extraction of Pr from Nd	**Pr**,Nd
b	Extraction of Ce from La	**Ce**,La

In the original text, the author of 'Art vs Science' structure commented that the recovery of MREO as well as HREO should be preferably outsourced, and hence, high attention is given solely on the LREO separation. However, the context of separation of this study is assumed to be conducted under one large operation, which covering all spectrums of REEs that available, particularly corresponding to the 'Art vs Science' process route.

5. METHODOLOGY

This section primarily explains the proposed methodology in obtaining as well as the procedural of analysing the SI outcomes. A number of work constraints are also employed in conjunction to the 'Art vs Science' structure that discussed previously as listed in the following:

i. The number of extraction as well as scrubbing stages are determined based on the concurrent SX operation that developed by Prof. Xu and his colleagues (Xu, Li, & Yian, 1985)
ii. HDEHP-HCL system (Sato, 1989) is used as the basis for extraction medium as well as the main SF reference.
iii. All calculations are developed based on Microsoft Excel platform.

Figure 3 denotes the complete procedures that conducted in this study which comprises of six (6) main stages. Each of the stages is provided with the corresponding schematic diagram of model that applied for the sake of explanation only. Nonetheless, no specific format restriction should be imposed whenever it is implemented for other cases (so long the principles are fully complied).

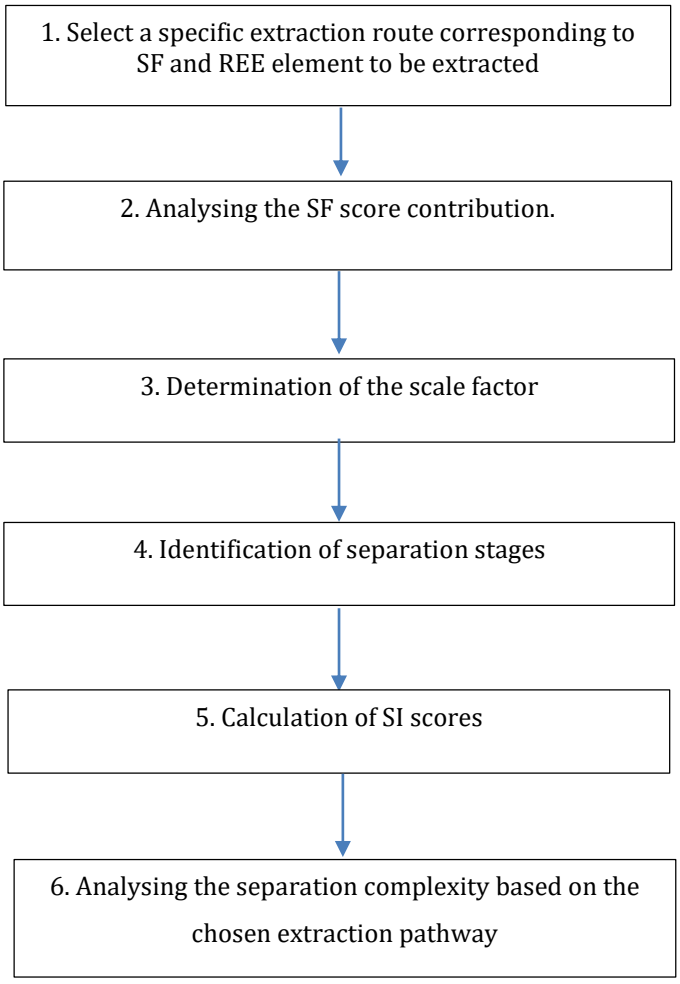


Figure 3: Procedures in calculating and analysing SI

In **step 1**, a specific extraction pathway such as 'Art vs Science' model is selected, which corresponding to the specified REE material that to be extracted. In this stage also, at least one or multiple sets of separation factors, whichever relevant, is/are applied concurrently as the main extractant to be analysed. Basically, this particular step focuses on consolidating all the essential information which formed as the basic foundation for analysis.

β	La	Ce	Pr	Nd	Sm	Eu	Gd	Tb	Dy	Ho	Er	Y
La	0	2.17	2.07	3.99	2.76	7.60	8.37	9.03	12.07	16.37	19.78	18.10
Ce	2.17	0.00	0.95	1.84	2.66	3.50	3.86	4.16	5.56	7.55	9.12	8.34
Pr	2.07	0.95	0.00	1.93	2.79	3.68	4.05	4.37	5.84	7.92	9.57	8.76
Nd	3.99	1.84	1.93	0.00	1.44	1.90	2.10	2.26	3.02	4.10	4.96	4.54
Sm	2.76	2.66	2.79	1.44	0.00	1.32	1.45	1.57	2.09	2.84	3.43	3.14
Eu	7.60	3.50	3.68	1.90	1.32	0.00	1.10	1.19	1.59	2.15	2.60	2.38
Gd	8.37	3.86	4.05	2.10	1.45	1.10	0.00	1.08	1.44	1.95	2.36	2.16
Tb	9.03	4.16	4.37	2.26	1.57	1.19	1.08	0.00	1.34	1.81	2.19	2.00
Dy	12.07	5.56	5.84	3.02	2.09	1.59	1.44	1.34	0.00	1.36	1.64	1.50
Ho	16.37	7.55	7.92	4.10	2.84	2.15	1.95	1.81	1.36	0.00	1.21	1.11
Er	19.78	9.12	9.57	4.96	3.43	2.60	2.36	2.19	1.64	1.21	0.00	0.91
Y	18.10	8.34	8.76	4.54	3.14	2.38	2.16	2.00	1.50	1.11	0.91	0

Figure 4: SF reference set based on HDEHP-HCL system (Sato, 1989)

In the second step (**step 2**), the main aim is to obtain the score contribution that resulted from the SF configuration that selected in step 1. For instance, Figure 2 denotes the set of SF reference that has been selected in this study which is presented using the matrix format.

β	La	Ce	Pr	Nd	Sm	Eu	Gd	Tb	Dy	Ho	Er	Y	
La	0	2.17	2.07	3.99	2.76	7.60	8.37	9.03	12.07	16.37	19.78	18.10	
Ce	2.17	0.00	0.95	1.84	2.66	3.50	3.86	4.16	5.56	7.55	9.12	8.34	
Pr	2.07	0.95	0.00	1.93	2.79	3.68	4.05	4.37	5.84	7.92	9.57	8.76	
Nd	3.99	1.84	1.93	0.00	1.44	1.90	2.10	2.26	3.02	4.10	4.96	4.54	
Sm	2.76	2.66	2.79	1.44	0.00	1.32	1.45	1.57	2.09	2.84	3.43	3.14	
Eu	7.60	3.50	3.68	1.90	1.32	0.00	1.10	1.19	1.59	2.15	2.60	2.38	
Gd	8.37	3.86	4.05	2.10	1.45	1.10	0.00	1.08	1.44	1.95	2.36	2.16	
Tb	9.03	4.16	4.37	2.26	1.57	1.19	1.08	0.00	1.34	1.81	2.19	2.00	
Dy	12.07	5.56	5.84	3.02	2.09	1.59	1.44	1.34	0.00	1.36	1.64	1.50	
Ho	16.37	7.55	7.92	4.10	2.84	2.15	1.95	1.81	1.36	0.00	1.21	1.11	
Er	19.78	9.12	9.57	4.96	3.43	2.60	2.36	2.19	1.64	1.21	0.00	0.91	
Y	18.10	8.34	8.76	4.54	3.14	2.38	2.16	2.00	1.50	1.11	0.91	0	
Total	102.31	49.71	51.93	32.08	25.49	29.01	29.92	31.00	37.45	48.37	57.77	52.94	547.98
ART VS SC STRUCTURE													
Step 1	9.30	4.52	4.72	2.92	2.32	2.64	2.72	2.82	3.40	4.40	5.25	4.81	4.15
	5.36				3.54								

Figure 5: SF reference set based on HDEHP-HCL system (Sato, 1989)

Next, the SF average of every REE ($\overline{SF_i}$) that involved in every individual extraction step is calculated as denoted in equation 2 as well as graphically presented in Figure 7.

$$\overline{SF_j} = \frac{\sum SF_i}{n_i - 1} \qquad (2)$$

Where, i = Every individual REE in REE 'j' column of Figure 6 according to the particular extraction step.

j = The main individual REE or group of interest that involved for extraction in that particular extraction step.

n_i = Total number of REE in that particular extraction step.

Then, the SF average of the individual REE is calculated according to the primary and alternative extraction element ($\overline{SF}_{p/a}$) correspond to $\overline{SF_j}$ value as shown in equation 3 as well as shown in Figure 5. The primary route (represented by symbol (i)) is based on the proposed extraction sequence that

suggested by (Mackowski, 2014), whereas the alternative routes (other than (i)) relate to other options of REE extraction which available for that particular separation step.

$$\overline{SF}_{p/a} = \frac{(\sum \overline{SF_j})_{p/a}}{n_{p/a}} \quad (3)$$

Where, $(\sum \overline{SF_j})_{p/a}$ = Summation of all $\overline{SF_j}$ of every REE that contained in either the primary *(p)* or alternative *(a)* group of that particular extraction step.

$n_{p/a}$ = total number of REE that contained in either the primary *(p)* or alternative *(a)* group of that particular extraction step.

The SF score is determined simply by obtaining the ratio of primary and alternative extraction element (R_p or R_a) over the SF average $\overline{SF_s}$ of that particular extraction step as indicated in equation 4 and also highlighted in Figure 6. From Figure 6, the diagram specifically referring to the particular extraction step 1 of 'Art vs Science' model.

$$R_{p/a} = \frac{\overline{SF}_{p/a}}{\overline{SF_s}} \quad (4)$$

Where, $\overline{SF_s} = \frac{\sum \overline{SF}_j}{n}$ and '*n*' = total number of all REEs.

ART VS SC STRUCTURE													Primary	Alternative
Step 1	9.30	4.52	4.72	2.92	2.32	2.64	2.72	2.82	3.40	4.40	5.25	4.81	4.15	
	5.36				3.54								0.854	1.292

R_p R_a

Figure 6: Position of *Rp* or *Ra* based on SF reference set

The final procedure of step 2 involves with the calculation of R_p and R_a for all the extraction steps which can be listed as depicted in Figure 7 once they are completed. In other words, equations 2 until 4 should be performed for every individual step of extractions within the process model that has been selected in step 1.

Steps	Separation Factor Score ($R_{p/a}$)
1 (i)	0.85
1 (ii)	1.29
2 (i)	1.04
2 (ii)	0.98
2a (i)	0.99
2a (ii)	0.94
2a (iii)	1.07
2b (i)	1.00
2b (ii)	1.00
2c (i)	-
2d (i)	0.91
2d(ii)	1.12
2d(iii)	0.92
2d(iv)	1.06
3 (i)	0.98
3 (ii)	1.02
3a(i)	1.00
3a(ii)	1.00
3b(i)	1.00
3b(ii)	1.00

Figure 7: *Rp* and *Ra* for all extraction steps of Art vs Sc structure

Step 3 basically relates to the determination of the scale factor which is derived based on the SP value and REE mineral characterization. In this work, the material specification is modified whereby a new average is calculated for every single REE. The composition distribution is determined for all the extraction steps respectively as shown in Figure 8.

No.	REE	Content in 150µg REE		Content by Category		Art vs Sc								
		Weight, µg	%	Weight, µg	%	1	2	2a	2b	2c	2d	3	3a	3b
1	La	30.71215	20.47%	128.21	85%	85%						73%		33%
2	Ce	63.01999	42.01%											67%
3	Pr	7.249437	4.83%									27%	21%	
4	Nd	27.22467	18.15%										79%	
5	Sm	5.126484	3.42%	10.68	7%	15%	49%	48%	83%					
6	Eu	1.043036	0.70%					10%	17%					
7	Gd	4.515414	3.01%					42%						
8	Tb	0.718453	0.48%	11.11	7%		51%			6.47%	6%			
9	Dy	4.06067	2.71%							36.55%	37%			
10	Ho	0.835138	0.56%							7.52%	8%			
11	Er	2.343557	1.56%							21.10%	21%			
12	Tm	0.35534	0.24%							3.20%	3%			
13	Yb	2.418608	1.61%							21.77%	22%			
14	Lu	0.377044	0.25%							3.39%	3%			
Total Weight, mg		150	100%	Total Weight, mg		150	22	11	6	11	11	128	34	94

Figure 8: REE composition distribution corresponding to all steps in the 'Art vs Science' extraction pathway

From Figure 8, almost all types of REEs are available excluding Sc and Y, and thus, the extraction of Y (step 2c) is removed from the analysis. Those composition values which highlighted in red are the particular REEs of interest that correspond to the description provided. The scale factor (F_{sf}) of every individual extraction step is computed based on the following equation 5 which involving both the primary as well as alternative routes. All F_{sp} values of every extraction step can then be placed in a single column as denoted in Figure 9. Those values that highlighted in red relate to the highest value of every separation step.

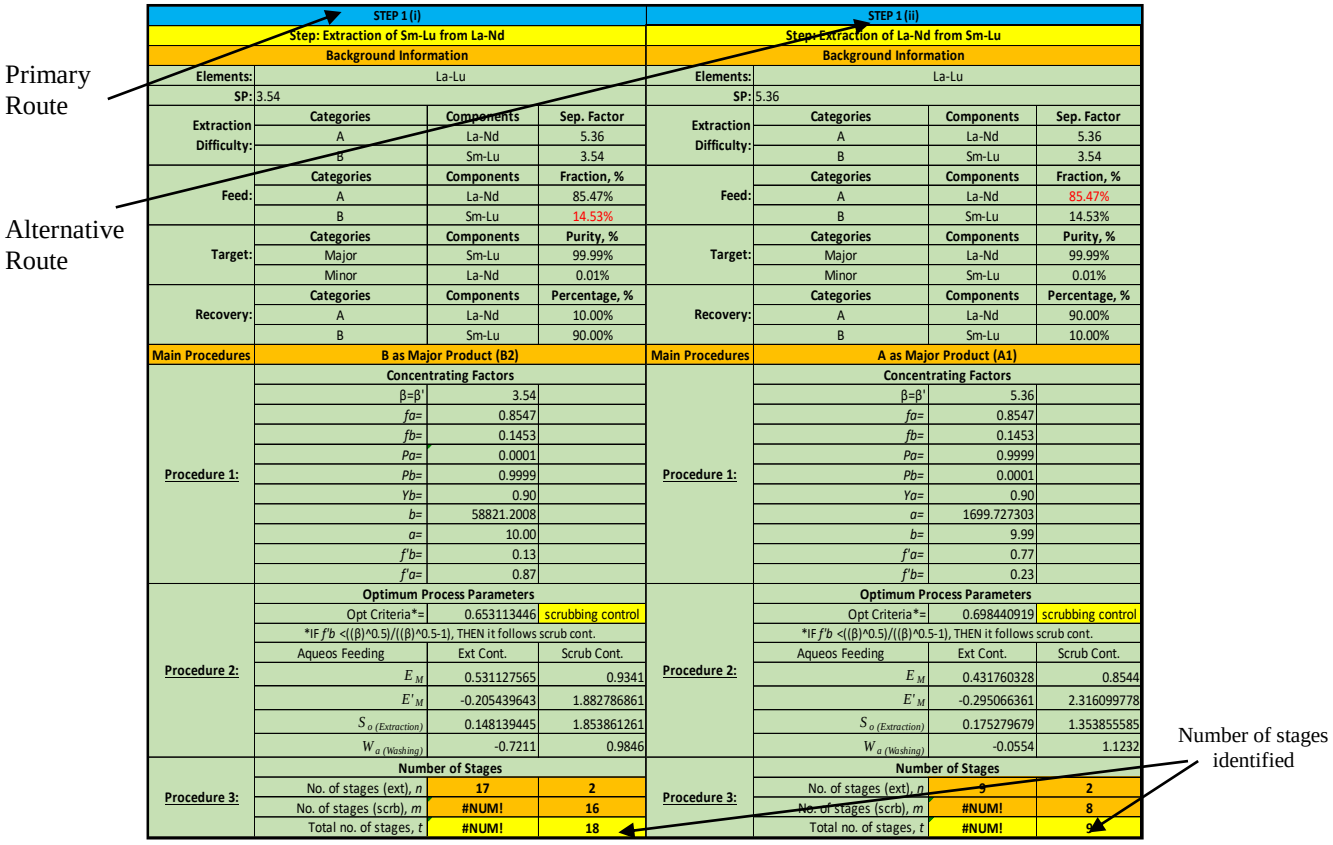
$$F_{sf} = R_{p/a} \times C \tag{5}$$

Where, C is obtained according to the corresponding REE characterization distribution as shown in Figure 8 previously.

Steps	Separation Factor Score	Scale Factor
1 (i)	0.85	12.41
1 (ii)	1.29	110.44
2 (i)	1.04	53.01
2 (ii)	0.98	47.85
2a (i)	0.99	41.77
2a (ii)	0.94	9.16
2a (iii)	1.07	51.51
2b (i)	1.00	83.09
2b (ii)	1.00	16.91
2c (i)	-	-
2d (i)	0.91	33.22
2d(ii)	1.12	7.23
2d(iii)	0.92	6.90
2d(iv)	1.06	22.27
3 (i)	0.98	26.39
3 (ii)	1.02	74.47
3a(i)	1.00	21.03
3a(ii)	1.00	78.97
3b(i)	1.00	67.23
3b(ii)	1.00	32.77

F_{sp}

Figure 9: The scale factor values for all extraction steps of 'Art vs Science' model



STEP 1 (i)				STEP 1 (ii)			
Step: Extraction of Sm-Lu from La-Nd				Step: Extraction of La-Nd from Sm-Lu			
Background Information				Background Information			
Elements:	La-Lu			Elements:	La-Lu		
SP:	3.54			SP:	5.36		
Extraction Difficulty:	Categories	Components	Sep. Factor	Extraction Difficulty:	Categories	Components	Sep. Factor
	A	La-Nd	5.36		A	La-Nd	5.36
	B	Sm-Lu	3.54		B	Sm-Lu	3.54
Feed:	Categories	Components	Fraction, %	Feed:	Categories	Components	Fraction, %
	A	La-Nd	85.47%		A	La-Nd	85.47%
	B	Sm-Lu	14.53%		B	Sm-Lu	14.53%
Target:	Categories	Components	Purity, %	Target:	Categories	Components	Purity, %
	Major	Sm-Lu	99.99%		Major	La-Nd	99.99%
	Minor	La-Nd	0.01%		Minor	Sm-Lu	0.01%
Recovery:	Categories	Components	Percentage, %	Recovery:	Categories	Components	Percentage, %
	A	La-Nd	10.00%		A	La-Nd	90.00%
	B	Sm-Lu	90.00%		B	Sm-Lu	10.00%
Main Procedures	B as Major Product (B2)			Main Procedures	A as Major Product (A1)		
Procedure 1:	Concentrating Factors			Procedure 1:	Concentrating Factors		
	β=β'	3.54			β=β'	5.36	
	fa=	0.8547			*fa*=	0.8547	
	fb=	0.1453			*fb*=	0.1453	
	Pa=	0.0001			*Pa*=	0.9999	
	Pb=	0.9999			*Pb*=	0.0001	
	Yb=	0.90			*Ya*=	0.90	
	b=	58821.2008			*a*=	1699.727303	
	a=	10.00			*b*=	9.99	
	f'b=	0.13			*f'a*=	0.77	
	f'a=	0.87			*f'b*=	0.23	
Procedure 2:	Optimum Process Parameters			Procedure 2:	Optimum Process Parameters		
	Opt Criteria*=	0.653113446	scrubbing control		Opt Criteria*=	0.698440919	scrubbing control
	*IF f'b <((β)^0.5)/((β)^0.5-1), THEN it follows scrub cont.				*IF f'b <((β)^0.5)/((β)^0.5-1), THEN it follows scrub cont.		
	Aqueos Feeding	Ext Cont.	Scrub Cont.		Aqueos Feeding	Ext Cont.	Scrub Cont.
	E_M	0.531127565	0.9341		E_M	0.431760328	0.8544
	E'_M	-0.205439643	1.882786861		E'_M	-0.295066361	2.316099778
	$S_{o\ (Extraction)}$	0.148139445	1.853861261		$S_{o\ (Extraction)}$	0.175279679	1.353855585
	$W_{a\ (Washing)}$	-0.7211	0.9846		$W_{a\ (Washing)}$	-0.0554	1.1232
Procedure 3:	Number of Stages			Procedure 3:	Number of Stages		
	No. of stages (ext), *n*	17	2		No. of stages (ext), *n*	9	2
	No. of stages (scrb), *m*	#NUM!	16		No. of stages (scrb), *m*	#NUM!	8
	Total no. of stages, *t*	#NUM!	18		Total no. of stages, *t*	#NUM!	9

Figure 10: Procedures in calculating stages based on step 1 (extraction of MREO and HREO from LREO)

The **fourth step** is the particular procedure that calculate the required number of separation stages, which include the extraction and scrubbing (or washing) operation, based on the specified REE of interest. As mentioned at the beginning, all the stages are calculated based on the procedures developed in Zhang *et al.,* 2016 (correspond to the equations). Figure 10 shows one example of stages calculation based on step 1 which involving the primary (extraction of MREO and HREO from LREO) and also alternative routes (extraction of LREO from MREO and HREO). All of these stages of every step are compiled in a single column as shown in Figure 11.

Steps	Separation Factor Score	Scale Factor	No. of Stages (Rough Estimation)
1 (i)	0.85	12.41	18
1 (ii)	1.29	110.44	9
2 (i)	1.04	53.01	35
2 (ii)	0.98	47.85	37
2a (i)	0.99	41.77	91
2a (ii)	0.94	9.16	132
2a (iii)	1.07	51.51	69
2b (i)	1.00	83.09	86
2b (ii)	1.00	16.91	64
2d (i)	-	-	-
2d (i)	0.91	33.22	60
2d(ii)	1.12	7.23	44
2d(iii)	0.92	6.90	67
2d(iv)	1.06	22.27	45
3 (i)	0.98	26.39	30
3 (ii)	1.02	74.47	23
3a(i)	1.00	21.03	35
3a(ii)	1.00	78.97	28
3b(i)	1.00	67.23	27
3b(ii)	1.00	32.77	29

Figure 11: The number of stages for all extraction steps of 'Art vs Science' model

Step 6 refers to the calculation of SI score that correlate to each separation phase that specified. As discussed in Section 3.0, the SI score is computed based on the ratio between the scale factor and number of stages as represented by equation 1 previously. All the calculated SI scores for every stage are placed in a single column as shown in Figure 12. Those values that highlighted in red are the highest scores corresponding to those separation steps respectively.

Steps	Separation Factor Score	Scale Factor	No. of Stages (Rough Estimation)	Separation Index *SI*
1 (i)	0.85	12.41	18	0.68
1 (ii)	1.29	110.44	9	11.79
2 (i)	1.04	53.01	35	1.52
2 (ii)	0.98	47.85	37	1.29
2a (i)	0.99	41.77	91	0.46
2a (ii)	0.94	9.16	132	0.07
2a (iii)	1.07	51.51	69	0.75
2b (i)	1.00	83.09	86	0.97
2b (ii)	1.00	16.91	64	0.26
2c (i)	-	-	-	-
2d (i)	0.91	33.22	60	0.56
2d(ii)	1.12	7.23	44	0.16
2d(iii)	0.92	6.90	67	0.10
2d(iv)	1.06	22.27	45	0.50
3 (i)	0.98	26.39	30	0.89
3 (ii)	1.02	74.47	23	3.17
3a(i)	1.00	21.03	35	0.60
3a(ii)	1.00	78.97	28	2.85
3b(i)	1.00	67.23	27	2.53
3b(ii)	1.00	32.77	29	1.14

Figure 12: The SI scores for all extraction steps of 'Art vs Science' model

Steps	Separation Factor Score	Scale Factor	No. of Stages (Rough Estimation)	Separation Index
1 (i)	0.85	12.41	18	0.68
1 (ii)	1.29	110.44	9	11.79
2 (i)	1.04	53.01	35	1.52
2 (ii)	0.98	47.85	37	1.29
2a (i)	0.99	41.77	91	0.46
2a (ii)	0.94	9.16	132	0.07
2a (iii)	1.07	51.51	69	0.75
2b (i)	1.00	83.09	86	0.97
2b (ii)	1.00	16.91	64	0.26
2c (i)	-	-	-	-
2d (i)	0.91	33.22	60	0.56
2d(ii)	1.12	7.23	44	0.16
2d(iii)	0.92	6.90	67	0.10
2d(iv)	1.06	22.27	45	0.50
3 (i)	0.98	26.39	30	0.89
3 (ii)	1.02	74.47	23	3.17
3a(i)	1.00	21.03	35	0.60
3a(ii)	1.00	78.97	28	2.85
3b(i)	1.00	67.23	27	2.53
3b(ii)	1.00	32.77	29	1.14
Total of Scores According to The Proposed Route	**8**	**338**	**381**	**0.89**
Total of Scores According to The Alternative Pathway	**9**	**460**	**337**	**1.37**

Figure 13: The overall analysis between the proposed and alternative separation pathway of 'Art vs Science' model

The final procedure (**step 6**) of the proposed methodology concerns on analysing the complexity of the chosen REE extraction pathway against the alternative route based on the SI score prioritization. As explained in section 3, SI is introduced as the basis particularly in critically choosing the most relatively feasible element to be recovered against the others. By principle, the larger the SI value, the more feasible that particular REE material to be extracted, corresponding to a specific extraction step that selected. This is simply due to the fact that SI depends on three main sources: the value of SF as well as the material characteristics (both are contributed as the scale factor, F_{SF}) and also the number of stages. In referring to equation 1, the larger the F_{SF} value it means either it has higher SF (higher SF contributes to easier to extract) or the particular REE element is relatively richer in terms of material composition (higher composition contributes to more economy advantage). It could also be caused by the number of stages that computed for the targeted REE element is relatively smaller compared to other REEs. In other words, the concept of 'feasibility' that applied in this work is defined as the sufficient amount of material to be recovered with relatively small number of stages as well as easily to be implemented.

In light of this concept, thus, the following procedures are proposed for conducting the separation complexity analysis.

i. Highlight as well as define the extraction option which having the highest SI value. From Figure 12, those extraction options which hold the highest SI value are highlighted in red (this has been performed according to every step respectively).
ii. Define the grand SI value of the primary extraction route, which consists of all the proposed extraction orders that promoted by the extraction structure that selected. From Figure 12, all the primary (proposed) routes are defined by (i) for every extraction step that applied. In obtaining the grand SI, the total of scale factors as well as number of stages are calculated, and followed by applying equation 1. As a result, the grand SI for the proposed pathway is 0.89.
iii. Define the grand SI value of the alternative extraction routes that own the highest SI value in every extraction step, which consists of both primary as well as other REE element extraction pathways. From Figure 12, all of these pathways are perceived as the more feasible or ideal extraction pathway preferably to be conducted as alternative compared to the primary processing routes as suggested initially.
iv. Critical evaluation should be conducted subsequently in comparing between the primary and alternative SI. The main aim of this procedure is to justify that the SI of alternative route is always showing the higher SI value, which offering lesser number of stages as well as higher scale factor (relatively higher recovery).

6. RESULTS AND DISCUSSION

The results of this work are discussed in three main segments. The first discusses on the separation complexity analysis based on the SF parameter alone as typically performed traditionally. Meanwhile, the second part explains the assessment of the REEs recovery solely on the scale factor which has been derived from the combination between SF and material characterization. While, the last segment focuses on the credibility of using the proposed SI index in evaluating the separation of REEs.

6.1. Prioritization of Separation Complexity Based on Separation Factor Score

As mentioned earlier, the main set of SFs that utilised in this study is based on HDEHP-HCL system. Figures 4 and 5 denote the original and total values (according to every element by column) of HDEHP-HCL SFs respectively, meanwhile the outcomes of applying the proposed methodology based on this system has been shown comprehensively in Figure 13 previously. Based on the total values of Figure 5, obviously the light REEs are relatively have greater amount compared to the medium or high REEs. This suggests that light REEs are relatively lesser complicated to extract by means of stages comparing to medium or heavy REEs. The most difficult components are the medium REEs, whereby the total SFs of every medium REES acquires generally less than 30 respectively. These values are relatively the lowest among of all other REEs. This indicates that the number of stages in recovering the medium REEs should be relatively greater than light or heavy REEs.

In order to assess the credibility of the chosen extraction pathway as well as the methodology that proposed, another set of SFs is analysed in this study. This second set of SFs is based on RE(III)-HCL-EHEHPA system (Bautista, 1995), which is shown in Figure 14. From Figure 14, all total values of SFs are identified to be much higher compared to the HDEHP-HCL system. This suggests that the number of stages of the second set system should be much lesser relative to the first. Nonetheless, all the SFs for MREEs are remained the lowest compared to either LREO or HREO.

Separation factors of rare earths in (RE(III)-HCL-EHEHPA (Bautista 1995)														
β	La	Ce	Pr	Nd	Sm	Eu	Gd	Tb	Dy	Ho	Er	Tm	Yb	Lu
La	0	1.30	1.42	1.67	3.33	6.52	9.52	22.50	36.40	93.90	117.00	156.00	175	199
Ce	1.30	0.00	1.09	1.28	2.57	5.02	7.36	17.30	28.00	72.30	90.50	120.00	135	152
Pr	1.42	1.09	0.00	1.17	2.35	4.59	6.72	15.80	64.20	66.00	82.70	110.00	123	140
Nd	1.67	1.28	1.17	0.00	2.00	3.94	5.74	13.50	21.80	56.30	70.50	93.70	105	119
Sm	3.33	2.57	2.35	2.00	0.00	1.96	2.87	6.74	10.90	28.20	35.30	46.80	52.6	59.5
Eu	6.52	5.02	4.59	3.94	1.96	0.00	1.46	3.45	6.39	14.40	18.00	24.00	26.9	30.4
Gd	9.52	7.36	6.72	5.74	2.87	1.46	0.00	2.35	3.81	9.82	12.30	16.30	18.3	20.7
Tb	22.50	17.30	15.80	13.50	6.74	3.45	2.35	0.00	1.62	4.18	5.23	6.95	7.81	8.83
Dy	36.40	28.00	64.20	21.80	10.90	6.39	3.81	1.62	0.00	2.58	3.23	4.29	4.82	5.45
Ho	93.90	72.30	66.00	56.30	28.20	14.40	9.82	4.18	2.58	0.00	1.25	1.66	1.87	2.11
Er	117.00	90.50	82.70	70.50	35.30	18.00	12.30	5.23	3.23	1.25	0.00	1.00	1.00	1.00
Tm	156.00	120.00	110.00	93.70	46.80	24.00	16.30	6.95	4.29	1.66	1.00	0.00	1.00	1.00
Yb	175.00	135.00	123.00	105.00	52.60	26.90	18.30	7.81	4.82	1.87	1.00	1.00	0.00	1.00
Lu	199.00	152.00	140.00	119.00	59.50	30.40	20.70	8.83	5.45	2.11	1.00	1.00	1.00	0.00
Total	**823.56**	**633.72**	**619.04**	**495.60**	**255.12**	**147.03**	**117.25**	**116.26**	**193.49**	**354.57**	**439.01**	**582.70**	**653.30**	**739.99**

Figure 14: SF reference set based on RE(III)-HCL-EHEHPA system (Bautista, 1995)

However, in comparing the two SF sets that employed in this study, despite the high value, the RE(III)-HCL-EHEHPA system is still struggling in separating REEs within their respective intra-group members. For instance, high number of stages are expected to be implemented in separating either La or Ce within the light REE group as their SF numbers are rather small. The same situation may also be observed when conducting the individual separation of REEs for medium group as well as among of the heavy components (all indicate comparatively small SF values). This trend can be also generally notified based on the HDEHP-HCL system. Hence, the larger SF values that identified in the second SF set are primarily contributed from the SF value among of the inter-group members such as between the light and heavy components or among medium and heavy elements. Meanwhile, the difference between the light and medium groups is seemed not that significant. In short, by reviewing the SFs values alone, one can expect that separation between the light and heavy or the medium and heavy groups can be performed in reduced complexity against the individual separation among of the intra-group members.

6.2. Prioritization of Separation Complexity Based on Scale Factor

The scale factor is computed based on the mathematical contribution between SF score and material composition distribution. As stated earlier, this study adopts the material characterization that reported in Zhang et al., 2014 which is shown in Figure 8 previously. Meanwhile, the complete results of calculating the separation stages based on the 'Art vs Sc' are depicted in Figure 13. From Figure 13, the total calculated amount of stages for the primary routes is 381, whereas the total stages of alternative routes is 337 (reduced by 12 %). It is also observed that the stages that computed for separating all the medium and heavy groups from the light REEs (step 1) are found to be the lowest (18 stages-primary, 9 stages-alternative). In addition, the stages in separating the individual medium component is found to be among of the largest (steps 2a and 2b) – 91 and 86 for the primary routes, while 69 and 64 for the alternative pathways respectively. Both of these observations justify the presumption that discussed previously, which is the separation complexity depends on the SF value contribution (the higher SF value, the lesser separation difficulty by means of extraction and scrubbing stages).

With regard to the scale factor contribution, Figure 13 indicates that half of the eight (8) primary routes [2(i), 2b(i), 2d(i), 3b(i)] are found to be highlighted in red, which ideally signifies that the potential capacities of those particular REEs are relatively high. Of this pathways, only two (2) routes are identified to have relatively lower stages [2(i) and 3b(i)] respectively. When analysing the original material characteristics, there are three (3) routes are identified significantly retain the

highest compositions [2b(i)-Sm, 2d(i)-Dy, 3b(i)-Ce] based on Figure 8, while the composition of medium REEs that extracted in 2(i) is slightly smaller in proportion (49%) compared to the heavy REEs group (51%). Therefore, through this finding it is learnt that the main contributors by means of scale factors particularly for 2b(i), 2d(i), 3b(i) are generated by the high material composition, while 2(i) is affected greatly by its SF value (the SF score of the medium REEs is slightly higher than the heavy REEs-refer to Figure 13).

Steps	Separation Factor Score, %	Scale Factor	No. of Stages (Rough Estimatio	Separation Index
1 (i)	0.82	12.41	6	1.96
1 (ii)	1.46	110.44	3	32.10
2 (i)	1.47	53.01	7	8.04
2 (ii)	0.80	47.85	9	5.60
2a (i)	1.03	41.77	29	1.44
2a (ii)	0.82	9.16	46	0.20
2a (iii)	1.15	51.51	25	2.08
2b (i)	1.00	83.09	35	2.39
2b (ii)	1.00	16.91	16	1.04
2c (i)	-	-	-	-
2d (i)	1.13	33.22	17	1.92
2d(ii)	1.79	7.23	14	0.52
2d(iii)	0.70	6.90	30	0.23
2d(iv)	0.66	22.27	30	0.73
2d(v)	0.82	2.62	27	0.10
2d(vi)	0.90	19.65	21	0.94
2d(vii)	1.00	3.39	22	0.15
3 (i)	0.98	26.39	87	0.30
3 (ii)	1.02	74.47	65	1.15
3a(i)	1.00	21.03	150	0.14
3a(ii)	1.00	78.97	118	0.67
3b(i)	1.00	67.23	78	0.87
3b(ii)	1.00	32.77	86	0.38
Total of Scores According to The Proposed Route	**8**	**338**	**409**	**0.83**
Total of Scores According to The Alternative Pathway	**10**	**460**	**347**	**1.32**

Figure 15: Assessment of extraction priority based on Art vs Sc structure (extraction medium: RE(III)-HCL-EHEHPA system & material characteristics: Zhang, etal, 2014)

On the other hand, Figure 15 shows the complete assessment of 'Art vs Science' model based on RE(III)-HCL-EHEHPA system as well as mineral characterization that reported. From Figure 15, the followings are observed:

i. Total number of stages of alternative pathway (347) is comparatively smaller against the proposed routes (409). This suggests that the separation complexity has been reduced significantly.

ii. The lowest separation stages are observed in step 1 (similar to the HDEHP-HCL system) – 6 and 3 stages for the primary and alternative routes respectively.

iii. In contrary to the former SF set, the second SF set indicates step 3a (extraction of Pr from Nd) is determined the most difficult as the total stages are the largest – 150 and 118 corresponding to the primary and alternative routes respectively.

iv. Interestingly, the similar process routes as identified previously in HDEHP-HCL system, which consists of steps 2(i), 2b(i), 2d(i), 3b(i) of the primary routes are found to have relatively large scale factor respectively. This means that the configuration of both SF sets are comparatively identical particularly based on these four (4) separation routes.

6.3. Prioritization of Separation Complexity Based on Separation Index

In further analyzing the previous observations, this study has realized that there are discrepancies when comparing the separation complexity based on either separation stages or scale factor individually. In other words, by looking alone on the separation stage numbers, the outcome could be misleading as low number of stages does not directly indicate high coverage of REEs can be

obtained. This is happened as the amount of stages is also affected by the proportion of REEs that available (not only depending on the SF values alone). The higher the proportion, the bigger number of stages is expected to apply. In contrary, the bigger SF values, then the stages become much lower. In light of this situation, hence, there are always situations where high value of SF is obtained while the composition of REEs is found relatively small. This will create complication in making decision particularly in prioritizing which REEs should be considered primarily for recovery.

For instance, the previous selected separation routes - steps 2(i), 2b(i), 2d(i), 3b(i) are revised again based on the results displayed in Figures 13 and 15. Out of these routes, only 2(i) and 3b(i) steps denote relatively lower number of stages respectively based on both SF sets as indicated in Figures 13 and 15. This suggests that the new SF set is specifically indicating only Ce (instead of Sm, Dy and Ce) is the crucial REE which can be feasibly extracted in lesser complexity (in terms of separation stages) against the other REEs based on the primary route of 'Art vs Science' hierarchy. Meanwhile, in the case of 2b(i) and 2d(i), both steps have seen relatively huge stages against to the alternative REEs that available corresponding to those steps. As if Sm and Dy are not favourable or perceived to be complex contrary to the other REEs.

In addressing the issue, thus, the concept of SI is introduced. The previous four separation routes [2(i), 2b(i), 2d(i), 3b(i)] are revisited again for comprehensive explanation. In all of those four routes of Figures 13 and 15, the corresponding SI values have been determined to be the largest relative to the other REEs that available respectively. This outcome directly suggests that the SI value has properly provided the correct mechanism in prioritizing the extraction complexity. In particular, there is no obvious complication involved in the cases of 2(i) and 3b(i) as the scale factors as well as separation stages are relatively high and also low respectively. Meanwhile, when considering the cases of 2b(i) and 2d(i), the analysis on both situations have induced confusing as both factors are in contradiction with each other as explained in the previous paragraph. Nonetheless, the SI value has balanced up fairly both of the contradictory factors by indicating Sm and Dy as the most feasible (highest SI value) as well as should be primarily prioritized during separation.

In addition, from the observation on both Figures 13 and 15 also, the findings of other separation steps including steps 1, 2a, 3 and 3a suggest that the SI values have sufficiently guided the separation sequence correctly. However, the separation on those routes should be applied based on the alternative pathways (SI values that highlighted in red based on Figures 13 and 15 instead of using the traditional 'Art vs Science' approach. In analysing the whole separation steps, both Figures 13 and 15 signify that the scale factor becomes the dominant criteria which affect the SI value greatly in contra to the separation stages number. This can be clearly evident by observing that the SI with the highest values are always corresponding the high scale factor value of the same row in Figures 13 and 15 respectively.

From both Figures 13 and 15 also, it is generally found that the former set of separation factor is the most feasible medium of extraction particularly for that REE composition of Zhang et al, 2014. This is indicated by higher overall SF value of HDEHP-HCL system (as in Figure 13) relative to the RE(III)-HCL-EHEHPA set (as in Figure 15) in both primary or alternative routes respectively. Figures 13 and 15 also denote that the scale factor of both SF sets are equal, nonetheless they are different by means of separation stages, whereby HDEHP-HCL system produces lower number of stages (337 stages based on alternative route as shown in Figure 13) against RE(III)-HCL-EHEHPA (347 stages based on alternative route as shown in Figure 15). This is indirectly reflects that the separation stages factor is also playing important role in affecting the magnitude of SI value, and thus, increasing or reducing the SI value as in the case of HDEHP-HCL or RE(III)-HCL-EHEHPA respectively.

7. CONCLUSION AND RECOMMENDATIONS

The results of this work has significantly justify that the separation index has properly evaluated the sequence of REE separation complexity based on the scopes that specified. The proposed parameter has sufficiently utilised the three main parameters of SX system– separation factors, material characterization and also separation stages as its main mathematical foundation, and hence, the credibility of its outcome is relatively high. Besides, the study also has demonstrated that the 'Art vs Science' separation model is practical for REE recovery, whereby both extractants that chosen are feasible mainly for inter-group separation instead of among of the intra-group members. The SI assessment can be utilised perhaps, during the conceptual design of developing the commercial REEs separation plant.

The study can be further extended by studying the effect of utilising SI in different material characterization contexts as well as the possible implication on economic advantage or any sustainability evaluation. It is also interesting to explore the effectiveness of the proposed parameter in various recovery rate condition especially towards optimizing the operation cost and also the effect in the other separation model.

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